# Octupolar order in the multiple spin exchange model on a triangular lattice

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We show how a gapless spin liquid with hidden octupolar order arises in applied magnetic field, in a model applicable to thin films of $^3$He with competing ferromagnetic and antiferromagnetic (cyclic) exchange interactions. Evidence is also presented for nematic — i.e. quadrupolar — correlations bordering on ferromagnetism in the absence of magnetic field.

The triangular lattice occupies a special place in the history of quantum magnetism. Although it is the simplest lattice to exhibit geometrical frustration, this still has highly nontrivial consequences — notably, that the Ising antiferromagnet (AF) on a triangular lattice remains disordered even in the limit of zero temperature. This fact led Anderson to suggest that the ground state of the spin-1/2 Heisenberg AF on a triangular lattice would be a gapless spin-liquid state with "resonating valence bond" (RVB) character [1]. While this conjecture has since been disproved [2], the idea of the RVB state continues to exert a strong hold on the popular imagination, and has motivated a search for spin-liquid states in a wide variety of two-dimensional spin models [3].

Lately, attention has been refocused on the triangular lattice by the discovery of a number of experimental realizations of a quasi-2D spin-1/2 magnets with triangular coordination. These include the organic $(BEDTTTF)_2Cu_2(CN)_3$ [4], the transition metal chloride $Cs_2CuCl_4$ [5], and solid films of $^3$He absorbed on graphite [6]. Of these, $^3$He films offer the most perfect realization of a spin-1/2 triangular lattice, but with an interesting twist — the nearest–neighbor interaction is *ferromagnetic* (FM) and competes with an AF 4-spin cyclic exchange [7]. As such, $^3$He on graphite is one of a number of new quasi-two dimensional systems to exhibit "frustrated ferromagnetism" [8, 9]. However it is unique in both its purity, and in the possibility of tuning the ratio of the competing interactions continuously simply by varying the density of $^3$He atoms. An additional strong motivation for understanding this system comes from the fact that, for a range of densities bordering on ferromagnetism, the magnetic ground state of $^3$He films is a fully gapless spin-liquid [10, 11].

Recently, we proposed that the competition between FM and AF interactions in frustrated spin systems could lead to a new kind of gapless spin-liquid state with hidden multipolar order bordering on the FM phase [12, 13, 14] (for related, earlier work see [15]). In this Letter we extend these ideas to a triangular lattice spin-1/2 model
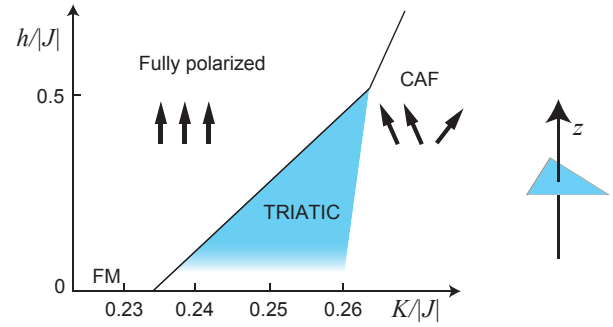


FIG. 1: (Color online). (Left) Schematic phase diagram for the spin-1/2 $J$-$K$ multiple spin exchange model on a triangular lattice in applied magnetic field, in the parameter range where $J < 0$ is FM and $K \geq 0$ is AF. A "triatic" phase with magnetic octupolar order appears in the shaded region between fully polarized and canted AF (CAF) phases. (Right) Schematic spin structure of this triatic order, which is invariant under spin rotations of $2\pi/3$ about the $z$ axis.

relevant to $^3$He on graphite [7]

$$\mathcal{H} = J \sum_{\langle ij \rangle} P_2 + K \sum_{\langle ijkl \rangle} (P_4 + P_4^{-1}) - h \sum_i S_i^z, \quad (1)$$

where $P_2$ and $P_4$ (cyclically) permute spins on nearest neighbor bonds $\langle ij \rangle$ and diamond plaquettes $\langle ijkl \rangle$, respectively. We include a coupling to magnetic field $h$, and consider exclusively FM $J < 0$ and AF $K > 0$.

Our main finding is that a dynamical process unique to 4-spin exchange on the triangular lattice leads to the formation of three magnon bound states which — under applied magnetic field — condense, giving rise to a new, partially-polarized quantum phase in which rotational symmetry is spontaneously broken, but without any long-range spin order perpendicular to the field (Fig. 1). We identify the order parameter for this novel phase as a rank three tensor with octupolar character, and confirm its existence through the exact diagonalization of finite-size clusters.

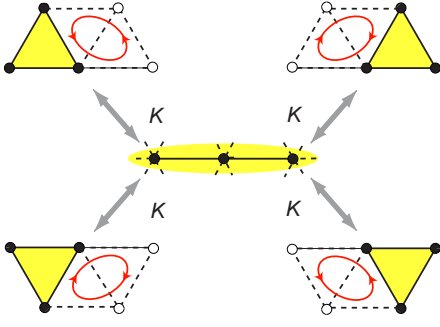To better understand the origins of this new quan-

FIG. 2: (Color online). The four-spin cyclic exchange operator can be used to propagate three flipped spins ($\bullet$) in a polarized FM background by tunneling between compact triangular and straight line configurations.
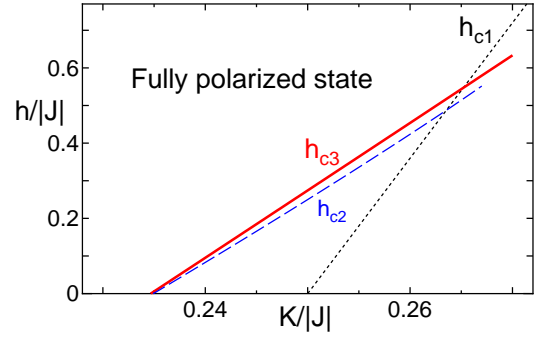


FIG. 3: (Color online). Critical fields $h_{c1}$ (dotted line), $h_{c2}$ (dashed line), and $h_{c3}$ (solid line), where one-, two-, and three-magnon instabilities destabilize the fully polarized state in the $J$-$K$ model with FM $J < 0$. The lines of $h_{c1}$ and $h_{c2}$ are given from exact solutions. The variationally estimated line $h_{c3}$ gives an exact lower bound for three-magnon instability line.

tum phase, let us first reconsider the classical $S \to \infty$ limit of Eq. (1). For $K/|J| < 1/4$, the ground state is a saturated FM. For $1/4 < K/|J| < 1$ it is highly degenerate [16]. Freely propagating domain walls destroy long range spin order, but nematic (i.e. quadrupolar) order persists [13]. This pathology is also evident in the spin excitations for the quantum spin-1/2 case. Within the FM phase, the one–magnon dispersion $\omega(\mathbf{q}) = -(J + 4K) \left[ 3 - \cos(q_x) - 2\cos(q_x)\cos(\sqrt{3}q_y/2) \right]$ is well defined. However exactly at the classical critical point $J = -4K$ it *vanishes* identically.

Nondispersing — i.e. localized — magnons are also found at the border of a FM phase in the equivalent multiple spin exchange model on the square lattice. In this case, pairs of flipped spins can propagate coherently under the action of AF spin exchange, even where a single magnon is localized. Magnons therefore form kinetic energy driven bound states, which condense and give rise to a quadrupolar "bond–nematic" order [14]. The energy of two flipped spins in a FM background can also be calculated *exactly* for the multiple spin exchange model on a triangular lattice. We find that two-magnon bounds states with total spin $S = N/2 - 2$ and momenta $\mathbf{q} = \{(0, 2\pi/\sqrt{3}), (\pi, \pi/\sqrt{3}), (-\pi, \pi/\sqrt{3})\}$ have a lower energy per spin than independent magnons for $K/|J| > 0.2349$.

However, the frustrated geometry of the triangular lattice introduces a new, two-step process by which a group of three flipped spins can move together — as shown in Fig. 2. This motivates us to try a simple trial wave function for a three spin bound state, within the restricted basis of states where the three spins form a compact straight line or triangle. Solving the Hamiltonian Eq. (1) in this restricted Hilbert space, we obtain five eigenmodes for the three-magnon bound states. The lowest eigenvalue, $E = -13J - 50K - \sqrt{J^2 + 4JK + 28K^2}$, corresponds to a state with momentum $\mathbf{q} = (0, 0)$, belonging to the trivial irrep of the space group, i.e. even under the space rotations $R_{2\pi/3}$, $R_\pi$ and the reflection $\sigma$. This variational

estimate of the three–magnon bound state is lower in energy than any single magnon or two–magnon state for a range of parameters $0.2347 < K/|J| < 0.2652$.

To put these calculations in a more physical context, let us consider the case in which a magnetic field $h$ is used to fully polarize the system. This field is then reduced until a critical value $h_c$ is reached, at which point the magnetization starts to change. We can calculate critical fields $h_{c1}$ and $h_{c2}$ at which one- and two-magnons start to Bose-condense. We can also estimate — using the trial wave function discussed above — the field $h_{c3}$ at which the system becomes unstable against the condensation of three-magnon bound states. In the absence of any other phase transition, the greatest of the three fields $h_{c1}$, $h_{c2}$ and $h_{c3}$ will determine the first instability of the system, and the nature of magnetic order near to saturation.

Performing this analysis, we find that the first instability for $K/|J| \gtrsim 0.27$ occurs in the one–magnon channel, and is against a three-sublattice canted AF, as previously found in mean field analysis [16]. However in the parameter range $0.2347 < K/|J| \lesssim 0.27$, $h_{c3} > h_{c2} \gg h_{c1}$ — see Fig. 3. Since the estimate of $h_{c3}$ is variational (while the values of $h_{c2}$ and $h_{c1}$ are exact), it provides a strict lower bound on $h_c$.

Exact diagonalization of finite clusters of up to $N = 36$ spins confirms that the transition out of the saturated state is controlled by three–magnon bound states for $0.24 \lesssim K/|J| \lesssim 0.28$. Results for a 36–spin cluster with $K/|J| = 0.25$ are shown in Fig. 4. Jumps of $|\Delta m| = 3$ are clearly visible in the magnetization process (inset) for fields close to saturation ($H_s \equiv h_{c3}$). The strong AF coupling $K$ ensure that interactions between three–magnon bound states are repulsive, and excitations containing more than three magnons therefore play no part in the transition. At finite $H_s$, the transition is of second order, and can be understood as a Bose–Einstein conden-
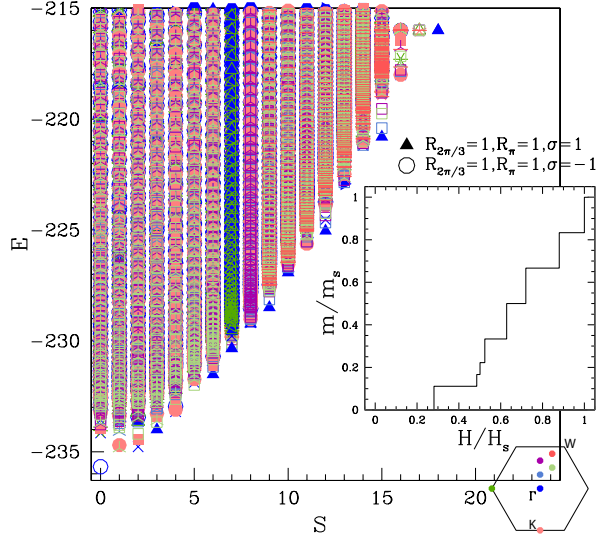
FIG. 4: Energy spectrum of the $J$-$K$ model for $N = 36$ spin cluster with $J = -4$ and $K = 1$, showing the oscillation in energies of the lowest lying states in spin sectors in the period three for large $S$. The inset shows the magnetization process $m/m_s$. Jumps of $|\Delta m| = 3$, corresponding to octupolar order, are clearly visible at large H. (Color online).

sation of three–magnon bound states. For $H_s \rightarrow 0$, this gives way to a first order transition.

These jumps can be traced to a set of states which lie at the bottom of spin sectors $S = 3n$, belonging to the trivial irrep of the space group. Each jump of $\Delta m = -3$ corresponds to adding one more three–magnon bound state to the system. The convex locus of these states as a function of $S$ implies that the transition at $h_{c3}$ is of second order. Numerical and analytic results close to saturation are therefore in perfect agreement. The condensation of three–magnon bound states will lead to a new form of quantum order. But what kind of order is it ?

We consider first the situation in applied magnetic field, where full $SU(2)$ spin symmetry is broken down to $U(1)$ rotations about the z-axis. In this case the order parameter is determined by the operator $O_{ijk}^{\dagger} = S_i^- S_j^- S_k^-$, which creates a three–magnon bound state in a polarized background. Both $\mathcal{R}e\ O^{\dagger}$ and $\mathcal{I}m\ O^{\dagger}$ are linear combinations of fully symmetrized rank-3 tensor operators

$$O_{ijk}^{\alpha\beta\gamma} = \sum_{a,b,c \in P(\alpha,\beta,\gamma)} S_i^a S_j^b S_k^c, \qquad (2)$$

and the resulting order parameter corresponds to a magnetic octupole on a triangular plaquette $\{ijk\}$.

As expected, both the real and the imaginary parts of the order parameter transform with period $2\pi/3$

$$\mathcal{R}e\ O_{ijk}^{\dagger} = \tfrac{4}{3}(O_{ijk}^{xxx} + R_{2\pi/3}^{\sigma}[O_{ijk}^{xxx}] + R_{4\pi/3}^{\sigma}[O_{ijk}^{xxx}]),$$

$$\mathcal{I}m\ O_{ijk}^{\dagger} = -\tfrac{4}{3}(O_{ijk}^{yyy} + R_{2\pi/3}^{\sigma}[O_{ijk}^{yyy}] + R_{4\pi/3}^{\sigma}[O_{ijk}^{yyy}]),$$

where $R_{\theta}^{\sigma}$ denotes a rotation in spin–space through angle $\theta$ about $z$ axis. The three–magnon condensate breaks the remaining $U(1)$ symmetry down to the symmetries of a triangle in the plane perpendicular to the applied field (Fig. 1). We therefore dub the resulting octupolar order "triatic". We note that triatic order of this type does *not* require any breaking of translational symmetry — the condensed three-magnon bound states can move, as long as they remain phase coherent. Triatic order of various types has previously been discussed in the context of the Heisenberg AF on a kagomé lattice [17].

Period–3 structure in the energy spectrum persists down to low spin in all of the clusters studied. However, new features appear in the structure of the spectrum for $S \lesssim 6$ (see Fig. 4). These features are sensitive to cluster geometry, which suggests that different types of order compete with octupolar order in the absence of magnetic field. While this makes interpretation of the spectra more difficult, it is nonetheless possible to place strong constraints on possible ground states.

First, it is possible to conclude that a new ground state exists between the FM phase and the short-ranged RVB spin liquid phase with finite spin-gap $\Delta \sim |J|$, which is known to occur for $K/|J| \approx 0.5$ [18]. For $0.235 \lesssim K/|J| \lesssim 0.28$ the ground state and low–lying excitations have different symmetries from the RVB phase [20]. Furthermore, in this parameter range, the rapid decay of excitation energies with system size suggests a gapless ground state for $h = 0$, and thus a breaking of $SU(2)$ symmetry.

Dipolar (Néel), quadrupolar (nematic) and octupolar (e.g. triatic) magnetic order all break $SU(2)$ symmetry, and each of these forms of order is associated with a specific "Anderson tower" of low–lying states [14]. The Anderson tower contains states in all spin sectors $S$ for Néel order; only states with even $S$ (period–2) for quadrupolar order, and only states in sectors with $S$ a multiple of three for *pure* octupolar order (period–3).

In view of the spectra, Néel order seems improbable. The number of low–lying states below the continuum in each spin sector $S$ is not compatible with what would be expected for dipolar order. Furthermore, a period–2 structure where the lowest lying states with odd $S$ are raised in energy relative to states with even $S$, is seen for $S \lesssim 6$ in those $N = 12$ and $N = 36$ spin clusters which possess the full symmetry of the lattice, and in the $N = 24$ cluster with the lowest ground-state energy. This period–2 structure suggests quadrupolar order is present in the ground state. The period–3 structure, present at larger $S$, is still clearly visible for small $S$ in some $N = 24$ clusters, but is hard to distinguish in the lowest–energy clusters. Moreover, the ground states for the $N = 12, 36$ clusters are not in the trivial irrep, which indicate that the symmetry of the ground state differs from the pure octupolar order observed in magnetic field.
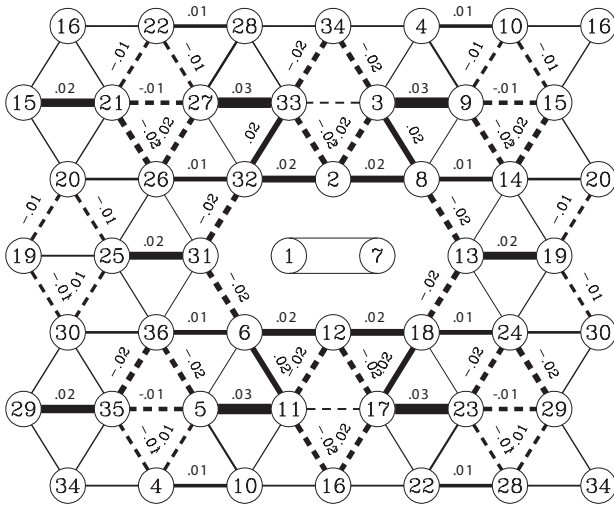
In fact, the symmetry and momenta of the low lying

FIG. 5: Bond nematic correlation in the ground state of the $J$-$K$ model for $N = 36$ spin cluster with $J = -4$, $K = 1$. (See Ref. [14] for the definition.) Correlations are measured relative to the reference bond 1–7. Positive (negative) correlations are drawn as full (dashed) lines. The thickness of the lines is a measure of the strength of the correlation.

even–$S$ states, and the corresponding quadrupolar correlation function (shown in Fig. 5), suggest the superposition of two distinct types of quadrupolar order. One corresponds to the condensation of s–wave magnon pairs with momenta at the $K$ points (e.g. $\mathbf{q} = (0, 2\pi/\sqrt{3})$) of the Brillouin zone — precisely the two–magnon instability of the saturated state predicted to occur at $h_{c2}$. The other corresponds to $d_{x^2-y^2}$+i$d_{xy}$–wave magnon pairs with momentum $\mathbf{q} = (0, 0)$.

We therefore conclude that the model has weak quadrupolar order in its ground state which extends from $K/|J| \approx 0.23$ to $K/|J| \approx 0.28$, where it undergoes a phase transition to a gapped RVB phase [21]. However in this parameter range, this gapless nematic phase lies close to the boundary with other competing phases.

We note that octupolar and quadrupolar orders are not exclusive: octupolar order is known to be accompanied by quadrupolar order in the ground state of spin $S = 3/2$ models with polynomial interactions [19]. This raises the question of the possibility of some similar coexistence in the present $S = 1/2$ model. Short-range FM correlations on individual triangular plaquettes ($\langle (\mathbf{S}_1 + \mathbf{S}_2 + \mathbf{S}_3)^2 \rangle = 2.42$ for $N = 36$, $K/|J| = 0.25$) persist within the ground state, reflecting the tendency to form three–magnon bound states. However, the present cluster sizes are too small to answer this question.

The *gapless* spin liquid state observed in $^3$He films occurs in the vicinity of the FM phase. It seems reasonable that a nematic phase (or octupolar phase) should interpolate between FM and RVB phases in the multiple spin exchange model on the triangular lattice, and that this state is therefore a strong contender to explain the spin–liquid seen in experiment.

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[20] We also note that the spectra showed no signature of the half–magnetization plateau known to occur in the RVB phase [16].
[21] It is also interesting to note that the gapful RVB wave function contains states with crossed singlet/triplet structure which is also seen in two–spin bound states near to saturation [13].